

Appln. No. 10/813,010
Appeal Brief

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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :
GOETZ-PETER SCHINDLER, ET AL : GROUP: 1624
SERIAL NO: 10/813,010 :
FILED: MARCH 31, 2004 : EXAMINER: SACKY, E.
FOR: PREPARATION OF AT LEAST ONE :
PARTIAL AND/OR
AMMOXIDATION PRODUCT OF A
HYDROCARBON

APPEAL BRIEF

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

SIR:

The following is an appeal to the Board of Appeals concerning the final decision by the Examiner to reject Claims 1-23 in the Official Action of June 2, 2008, concerning the above-identified application.

REAL PARTY IN INTEREST

BASF SE is the real party of interest in the above-identified application.

RELATED APPEALS AND INTERFERENCES

There are no other applications either on appeal or in interference that are related to the present application.

STATUS OF CLAIMS ON APPEAL

Claims 1-23 are pending and on appeal in the application.

STATUS OF AMENDMENTS

No amendment was filed in response to the final Office Action of June 2, 2008.

SUMMARY OF CLAIMED SUBJECT MATTER

The present invention is directed to a process for preparing at least one partial oxidation and/or ammoxidation product of hydrocarbon by subjecting at least one saturated hydrocarbon H to heterogeneously catalyzed dehydrogenation in the gas phase to form a product gas mixture A which comprises at least one partially dehydrogenated hydrocarbon H, leaving constituents present in the product gas mixture A, other than the saturated hydrocarbon H and other than the partially dehydrogenated hydrocarbon H therein, or partly or fully removing them to obtain a product gas mixture A'. The product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, is subjected to at least one heterogeneously catalyzed partial oxidation and/or ammoxidation of the at least one partially dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A'. The key feature of the present process is that the product gas mixture A, the product gas mixture A' and/or the gas mixture B is subjected to at least one mechanical separating operation by which solid particles present in these gas mixtures are removed before the at least one heterogeneously catalyzed partial oxidation and/or ammoxidation.

Support for the invention as summarized can be found in the paragraph of page 3, lines 15-29 of the specification.

GROUND OF REJECTION TO BE REVIEWED ON APPEAL

Whether Claims 1-23 stand properly rejected based on 35 USC 103(a) as obvious over Maier et al, EP 0938 463 in view of Maunders et al, U. S. Patent 5,550,309.

ARGUMENT

Rejection of Claim 1 over Maher et al and Maunders et al

Claim 1

The context of the discovery of the present invention is the well known procedure of preparing an at least one partial oxidation and/or ammoxidation product of a hydrocarbon by *first* subjecting at least one saturated hydrocarbon (H) to a heterogeneously catalyzed *dehydrogenation* in the gas phase to form a product gas mixture (A) which is comprised of at least one partially dehydrogenated hydrocarbon (H), as well as components other than saturated hydrocarbon (H) molecules and other than the partially dehydrogenated hydrocarbon (H) molecules. The components that are different from the partially dehydrogenated hydrocarbon (H) and the saturated hydrocarbon (H) can be partly or completely removed from the gas mixture (A) to form a gas mixture (A'). Gas mixture (A) and/or gas mixture (A') are used to form a gas mixture (B), and then in a second stage the resulting mixture (B) is subjected to partial oxidation and/or ammoxidation in a heterogeneously catalyzed reaction. The *second* stage catalyst which promotes the partial oxidation and/or ammoxidation of the unsaturated hydrocarbon (H) is of a substantially different type from the dehydrogenation catalyst of the first dehydrogenation stage mentioned above.

However, after some long term observations in-house of this process, applicants have found that very fine particles of dehydrogenation catalyst of the *first* are conveyed from the dehydrogenation zone into the subsequent zone where heterogeneously catalyzed partial oxidation and/or ammoxidation of the dehydrogenated hydrocarbon occurs. In fact, at least some of the fine *first* stage catalyst particles settle into the fixed catalyst employed in the *second* oxidation or ammoxidation zone, where, in the presence of excess oxygen, the fine catalyst particles can promote the undesired combustion of hydrocarbons to CO₂ and H₂O, as well as the reaction of hydrogen gas with oxygen to form water. These reactions are

undesirable and disadvantageous, since they can result in undesired reactant consumption during the partial oxidation or ammoxidation of dehydrogenated hydrocarbon to other than the desired products and other risks which can not be precisely identified beforehand such as corrosion of the apparatus employed in the process. This discovery then has led to the present invention which is that the removal of very fine particles of dehydrogenation catalyst from gas mixture (A) or gas mixture (A') before these gas mixtures enter the zone in which they are partially oxidized and/or ammoxidized results in higher reaction efficiency. Thus, the problems which attend the presence of very fine dehydrogenation catalyst particles in the oxidation or ammoxidation zone are avoided.

Appellants refer to the examples of the specification as a basis for demonstration of the effect of the present invention. The description of the experimental plant (Part 3) on pages 15 and 16 of the text describes the passage of LPG through a section made-up of a metering unit, a reactor unit, a cycle gas unit and a discharge unit. The passage of LPG containing gases through the section which employs the dehydrogenation catalyst is described. The experimental plant was operated in a series of cycles continuously over a period of three months under the conditions stated at the bottom of page 16. As described on page 17 of the text, at the end of the experimental series, flash rust was detected in considerable amounts in all parts of the experimental plant. An analysis of the rust showed that in addition to showing that the expected components of Fe, Ni and Cr (from the steel of the units), amounts of Zr, Si and Pt, which only could originate from dehydrogenation catalyst, were present. This evidence, which is not mere argument or conclusionary statements by appellants (O. A., June 2, 2008, page 3), therefore gave rise to the present invention which is that the removal of dehydrogenation catalyst from the gaseous effluent passed from the dehydrogenation section would be expected to provide material benefit in the overall conversion of saturated

hydrocarbon to oxidation and/or ammoxidation product in the two sequential stages of dehydrogenation and oxydehydrogenation each over a different heterogeneous catalyst.

As stated previously on the record concerning the cited and applied '463 reference, while it clearly discloses the partial oxidation of an unsaturated light hydrocarbon such as propylene over a heterogeneous catalyst to form a partially oxidized product such as acrolein in paragraph [0024] and succeeding paragraphs, nevertheless, the reference discloses that the starting unsaturated hydrocarbon is produced by a process of oxydehydrogenation of a starting saturated hydrocarbon such as propane over an oxydehydrogenation catalyst such as one of those disclosed in paragraphs [0019] and [0020]. Of critical importance is that there is no description or suggestion of a heterogeneously catalyzed dehydrogenation of a saturated hydrocarbon that occurs with the accompanying formation of molecular hydrogen as the primary by-product over a dehydrogenation catalyst. In the Office Action of February 27, 2007, the Examiner states in paragraph 3 on page 4 of the Office Action that in applicants' claimed process for preparing at least one partial oxidation and/or ammoxidation product of propylene, crude propane is subjected to a homogeneously and/or heterogeneously catalyzed dehydrogenation and/or oxydehydrogenation reaction. This is incorrect. The dehydrogenation of a saturated hydrocarbon such as propane in applicants' process does not occur by oxydehydrogenation (with water as the primary by-product), but by catalytic dehydrogenation with molecular hydrogen as the primary by-product. A dehydrogenation catalyst is used to catalyze the dehydrogenation reaction (a type of catalyst not used to promote oxydehydrogenation reactions), and it is this dehydrogenation catalyst that results in the formation of the very fine catalyst particles that previously had been conveyed from the dehydrogenation zone to the subsequent or following oxidation zone where unsaturated hydrocarbon (propene) is oxidized to the partially oxidized hydrocarbon (acrolein). Even in the case of oxydehydrogenation, as the first stage small particles of the oxydehydrogenation

subsequent or following oxidation zone where unsaturated hydrocarbon is partially oxidized, such particles would not cause the problems dehydrogenation catalyst particles cause in said zone (the underlying catalytic mechanism is different). Consequently, the '463 reference does not address appellants' solid particle removal procedure. Moreover, there is no teaching or suggestion in the reference that patentees ever contemplate the catalytic dehydrogenation of a saturated hydrocarbon, but only disclose catalytic oxydehydrogenation. It is therefore evident that the reference could not and does not teach the removal of very fine particles of dehydrogenation catalyst from a product stream comprised of newly or freshly formed unsaturated hydrocarbon (propene).

In citing the Maunders et al patent, the Examiner does so as evidence that the removal of catalyst particles from a process stream is routine. However, this is not what the reference describes, besides which the context of the reference is entirely different from the present invention. Maunders et al achieves its objective of the dehydrogenation of an alkane over a dehydrogenation catalyst by placing a second catalyst in the catalyst bed for the production of a dehydrogenated hydrocarbon product which functions as a hydrogen adsorbent and/or as an agent which reacts with adsorbed hydrogen. There is absolutely no description or suggestion of a two stage process in which a saturated hydrocarbon starting material is dehydrogenated over a first type of heterogeneous catalyst and then passing the gas mixture obtained, which contains some dehydrogenated hydrocarbon, over a heterogeneous oxidation catalyst where at least partial oxidation of ammoxidation occurs. Thus, Maunders et al not only is difficult to combine with '463, but also in describing a hydrogen adsorption mechanism in no manner speaks to the separation of dehydrogenation catalyst particles from a gas stream which is about to be subjected to partial oxidation and/or ammoxidation. Thus, the combined references are not believed to suggest the present invention.

The Examiner states that the essential process feature of the present claims of employing a mechanical means of separation of residual dehydrogenation catalyst from a gaseous unsaturated hydrocarbon containing intermediate is well known and therefore obvious. However, neither of the cited references or the background relevant prior art discussed in the present specification provides a context for an interruption of a two stage hydrocarbon conversion process in which entrained heterogeneous catalyst particles from a first stage hydrocarbon dehydrogenation are separated by some mechanical means before the gas phase containing dehydrogenated hydrocarbon is subjected to oxidation and/or ammoxidation over a heterogeneous catalyst specific for this type of reaction. The present invention as set forth in Claim 1 is believed to be patentable over the prior art of record.

Claims 2-23

The Examiner has placed all 23 of the active claims in one group. Arguments for Claims 2-23 as a group are now advanced, because they all share the common feature of being an aspect of a process of preparing at least one partial oxidation and/or ammoxidation product of a hydrocarbon by initially dehydrogenating a saturated hydrocarbon (H) over a heterogeneous dehydrogenation catalyst to at least one partially dehydrogenated hydrocarbon, and then subjecting the dehydrogenated hydrocarbon to an oxidation and/or ammoxidation over a heterogeneous catalyst suitable for this reaction. Of course, the process of the present invention is distinguished by the fact that the gas product produced by the initial dehydrogenation of the hydrocarbon is subjected to a mechanical separation operation in which dehydrogenation catalyst of the first stage entrained in the gas exiting the dehydrogenation zone is removed prior to passage of the at least partially unsaturated hydrocarbon into the zone in which oxidation and/or ammoxidation of the unsaturated hydrocarbon occurs. All of the dependent claims describe more specific details as to starting

materials and oxidation products produced. On the other hand, as stated above, Maier et al '463 discloses the initial reaction of a vaporous hydrocarbon such as propane by oxydehydrogenation to propene followed by conversion to aldehyde or acid product over an oxidation catalyst. This is not the process of the present invention. Maunders et al is even of lesser relevance to the invention than '463, because the reference only discloses the presence of a second catalyst in a catalyst bed whose function is to act as an absorber of hydrogen that is obtained by the catalytic dehydrogenation of an alkane reactant. Clearly, the dependent claims are patentable over the cited and applied prior art.

In view of the arguments set forth above, it is believed that the decision by the Examiner to continue the rejection of the claims is erroneous and should be REVERSED.

Respectfully submitted,

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CLAIM APPENDIX

Claim 1. A process for preparing at least one partial oxidation and/or ammoxidation product of hydrocarbon by subjecting at least one saturated hydrocarbon H to heterogeneously catalyzed dehydrogenation in the gas phase to form a product gas mixture A which comprises at least one partially dehydrogenated hydrocarbon H, leaving constituents present in the product gas mixture A, other than the saturated hydrocarbon H and other than the partially dehydrogenated hydrocarbon H therein, or partly or fully removing them to obtain a product gas mixture A', and subjecting product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, to at least one heterogeneously catalyzed partial oxidation and/or ammoxidation of the at least one partially dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A', which comprises subjecting the product gas mixture A, the product gas mixture A' and/or the gas mixture B, before the at least one heterogeneously catalyzed partial oxidation and/or ammoxidation, to at least one mechanical separating operation by which solid particles present in these gas mixtures are removed.

Claim 2. A process as claimed in claim 1, wherein the saturated hydrocarbon H is propane, and the heterogeneously catalyzed partial oxidation of the partially dehydrogenated hydrocarbon H is the partial oxidation of propene to acrolein and/or acrylic acid.

Claim 3. A process as claimed in claim 1, wherein the saturated hydrocarbon H is isobutane, and the heterogeneously catalyzed partial oxidation of the partially dehydrogenated hydrocarbon H is the partial oxidation of isobutene to methacrolein and/or methacrylic acid.

Claim 4. A process as claimed in claim 1, wherein the saturated hydrocarbon H is propane, and the heterogeneously catalyzed partial ammoxidation of the partially dehydrogenated hydrocarbon H is the partial ammoxidation of propene to acrylonitrile.

Claim 5. A process as claimed in claim 1, wherein the saturated hydrocarbon H is isobutane, and the heterogeneously catalyzed partial ammoxidation of the partially dehydrogenated hydrocarbon H is the partial ammoxidation of isobutene to methacrylonitrile.

Claim 6. A process as claimed in claim 1, wherein constituents present in the product gas mixture A, other than the saturated hydrocarbon H and other than the partially dehydrogenated hydrocarbon H therein, are partly or fully removed to obtain a product gas mixture A'.

Claim 7. A process as claimed in claim 1, comprising subjecting product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, to at least one heterogeneously catalyzed partial oxidation of the at least one partially dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A'.

Claim 8. A process as claimed in claim 1, comprising subjecting product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, to at least one heterogeneously catalyzed partial ammoxidation of the at least one partially dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A'.

Claim 9. A process as claimed in claim 1, comprising subjecting product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, to at least one heterogeneously catalyzed partial oxidation and ammoxidation of the at least one partially

dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A'.

Claim 10. A process as claimed in claim 6, comprising subjecting product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, to at least one heterogeneously catalyzed partial oxidation of the at least one partially dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A'.

Claim 11. A process as claimed in claim 6, comprising subjecting product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, to at least one heterogeneously catalyzed partial ammoxidation of the at least one partially dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A'.

Claim 12. A process as claimed in claim 6, comprising subjecting product gas mixture A and/or product gas mixture A', as a constituent of a gas mixture B, to at least one heterogeneously catalyzed partial oxidation and ammoxidation of the at least one partially dehydrogenated hydrocarbon H present in the product gas mixture A and/or product gas mixture A'.

Claim 13. A process as claimed in claim 7, wherein the saturated hydrocarbon H is propane, and the heterogeneously catalyzed partial oxidation of the partially dehydrogenated hydrocarbon H is the partial oxidation of propene to acrolein and/or acrylic acid.

Claim 14. A process as claimed in claim 7, wherein the saturated hydrocarbon H is isobutane, and the heterogeneously catalyzed partial oxidation of the partially

dehydrogenated hydrocarbon H is the partial oxidation of isobutene to methacrolein and/or methacrylic acid.

Claim 15. A process as claimed in claim 8, wherein the saturated hydrocarbon H is propane, and the heterogeneously catalyzed partial ammoxidation of the partially dehydrogenated hydrocarbon H is the partial ammoxidation of propene to acrylonitrile.

Claim 16. A process as claimed in claim 8, wherein the saturated hydrocarbon H is isobutane, and the heterogeneously catalyzed partial ammoxidation of the partially dehydrogenated hydrocarbon H is the partial ammoxidation of isobutene to methacrylonitrile.

Claim 17. A process as claimed in claim 10, wherein the saturated hydrocarbon H is propane, and the heterogeneously catalyzed partial oxidation of the partially dehydrogenated hydrocarbon H is the partial oxidation of propene to acrolein and/or acrylic acid.

Claim 18. A process as claimed in claim 10, wherein the saturated hydrocarbon H is isobutane, and the heterogeneously catalyzed partial oxidation of the partially dehydrogenated hydrocarbon H is the partial oxidation of isobutene to methacrolein and/or methacrylic acid.

Claim 19. A process as claimed in claim 11, wherein the saturated hydrocarbon H is propane, and the heterogeneously catalyzed partial ammoxidation of the partially dehydrogenated hydrocarbon H is the partial ammoxidation of propene to acrylonitrile.

Claim 20. A process as claimed in claim 11, wherein the saturated hydrocarbon H is isobutane, and the heterogeneously catalyzed partial ammoxidation of the partially dehydrogenated hydrocarbon H is the partial ammoxidation of isobutene to methacrylonitrile.

Claim 21. A process as claimed in claim 1, wherein the dehydrogenation catalyst is at least one metal deposited on a support.

Claim 22. A process as claimed in claim 1, wherein the catalytic dehydrogenation is conducted under a working pressure ranging from 0.3 to 3 atm

Claim 23. A process as claimed in claim 22, wherein the catalytic dehydrogenation is conducted in the presence of steam.

EVIDENCE APPENDIX

Appellants do not reply upon any evidence submitted pursuant to §§ 1.130, 1.131, or 1.132 of this title or any other evidence entered by the Examiner.

RELATED PROCEEDINGS APPENDIX

Appellants have no copies of decisions rendered by a court or the Board in any related case.